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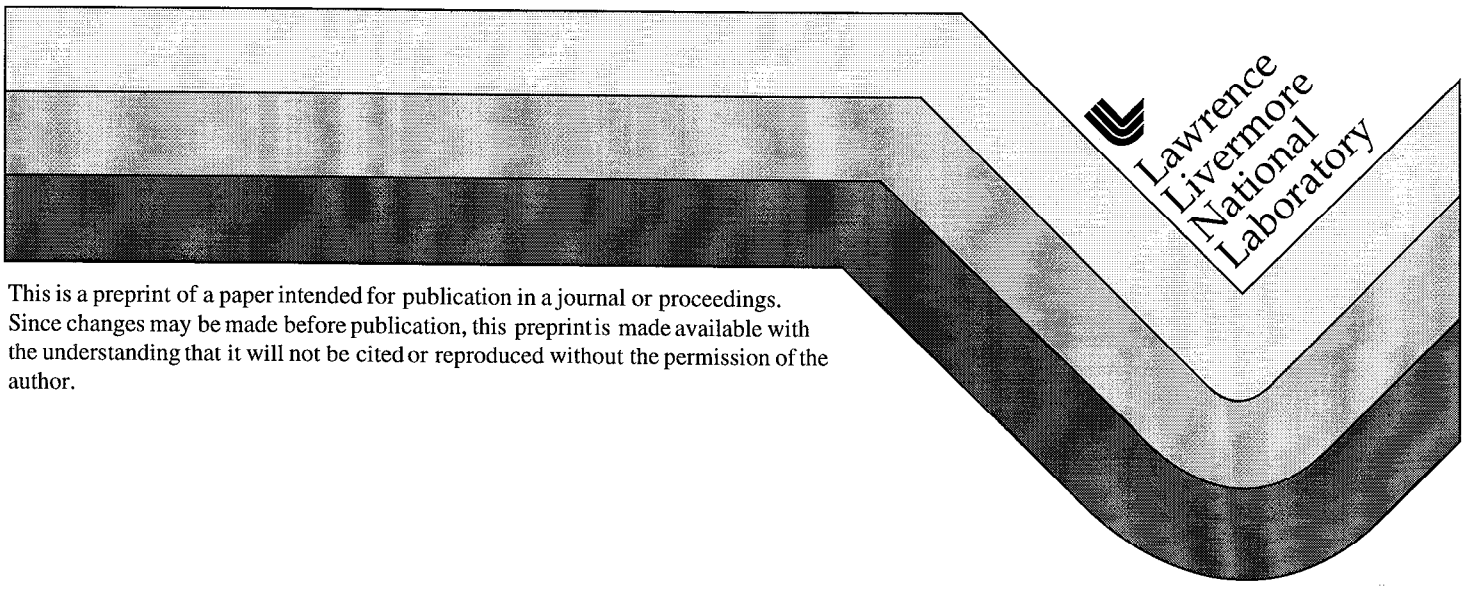

THE SPENT FUEL STANDARD - DOES THE CAN-IN-CANISTER CONCEPT FOR PLUTONIUM IMMOBILIZATION MEASURE UP?

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ABSTRACT

Critics continue to question whether or not the can-in-canister concept for immobilization and disposal of surplus plutonium meets the "'Spent Fuel Standard." Following this standard would make this plutonium roughly as "inaccessible for weapons use as the much larger and growing quantity of plutonium that exists in spent fuel from commercial reactors." These critics take a narrower view of the "Spent Fuel Standard" than was intended in the National Academy reports, rather than considering the total effective barrier. This paper directly compares retrieval and recovery of plutonium from a can-in-canister to a spent fuel assembly. The conclusion from this study, as from earlier studies, is that the plutonium in the can-in-canister form is less accessible and less attractive to a potential proliferate than the plutonium that exists in spent fuel from commercial reactors.

I. INTRODUCTION

With the fall of the Berlin Wall and the dissolution of the Soviet Union, the Cold War's nuclear arms race came to an abrupt halt. The United States and Russia began dismantling tens of thousands of nuclear weapons, resulting in large quantities of fissile materials becoming excess to military needs. In the U. S. National Academy of Sciences (NAS) Studies, [1,2] the Academy deemed the growing stockpile of excess fissile materials a "clear and present danger."

About 100 tonnes of plutonium will be removed from the US and Russian stockpiles[3-8] by the year 2005. For comparison, 1200 to 1600 tonnes of weapons-usable plutonium will exist in spent fuel from domestic and foreign commercial reactors by the year 2000. In the United States, spent fuel has been accumulating in storage pools since 1968 (Most of the fuel assemblies discharged from reactors prior to 1972 was processed.). At the midpoint of the immobilization program, about 2012, over 1800 fuel assemblies containing more than 400 tonnes of uranium will be over forty years old in the United States. Another 28,700 assemblies containing about 7200 tonnes of uranium will be between 30 and 40 years of age. The older spent fuel was irradiated to less burn-up than the younger fuel; therefore their fission product inventory is less. It is not anticipated that spent fuel will be shipped to the Federal Repository until after 2015. At the time shipment begins, the average, unweighted age of spent fuel will be about 24 years. At the midpoint of shipping the spent fuel, the average age will be approaching 30 years.

The NAS Committee on International Security and Arms Control stated[1,2] "while it ["reactor-grade" plutonium] could be used to make nuclear bombs, it poses much smaller risks than separated plutonium . . . because of the mass, bulk, and intense radiation field of the spent fuel assemblies and because of the additional technical sophistication and resources required for chemical separation . . . from the accompanying fission products and uranium."

The NAS further stated[1,2], "Options that left the weapons plutonium more accessible would mean that this material would continue to pose a unique safeguards problem indefinitely. Conversely, the costs, complexities, risks, and delays of going beyond the spent fuel standard to eliminate the excess weapons plutonium completely or nearly so would not offer substantial additional security benefits unless society were prepared to take the same approach with the global stock of civilian plutonium." Therefore, the principal goal is to render surplus plutonium as inaccessible and unattractive for reuse in nuclear weapons as it is in average spent reactor fuel. At the midpoint of shipping the high level waste canisters from Savannah River to the

Federal Repository, about 2020 or later, radiation from the immobilization product will compare favorably with 30 year old spent fuel.

The NAS spent nuclear fuel standard[1,2] does not actually require plutonium to be transformed into spent fuel. Likewise, the weapons-grade plutonium disposition product need not have all the characteristics of spent fuel to meet the objectives of the spent fuel standard. The idea behind the spent fuel standard is to create a variety of barriers to the recovery, the sum of which make it roughly as inaccessible and unattractive as recovery of plutonium from average spent fuel.

II. USEFULNESS OF VARIOUS GRADES OF PLUTONIUM IN A WEAPON

A paper by J. Carson Mark[9], former Director of the Theoretical Division of Los Alamos, discusses the question as to whether a bomb could be made from reactor grade plutonium. He reaches the following conclusions about the usefulness of plutonium in weapons:

- Reactor-grade plutonium with any level of irradiation is a potential explosive material.
- The difficulties of developing an effective design of the most straightforward type are not appreciably greater with reactor-grade plutonium than those that have to be met the use of weapons-grade plutonium.
- Hazards of handling reactor-grade plutonium, though somewhat greater than those associated with weapons-grade plutonium, are of the same type and can be met by applying the same precautions. This, at least, would be the case when fabricating a modest number of devices. For a project requiring an assembly line type of operation, more provisions for remote handling procedures for some stages of the work might be required that would be necessary for handling weapons-grade material or for handling a limited number of items.
- The need for safeguards to protect against the diversion and misuse of separated plutonium applies essentially equally to all grade of plutonium.

In short, the design of a crude nuclear explosive using higher burnup plutonium will have to account for the extra heat generation and radiation exposure, but provisions can be devised to cope with these features.

III. WHAT IS AVERAGE SPENT FUEL?

One hundred twenty-four types of reactor fuel assemblies have been used in 117 light water reactors (LWR) within the United States[10]. They may be broadly grouped as Boiling Water Reactors (BWR) and Pressurized Water Reactors (PWR). Between 1968 and 1992 about 37% by weight of the spent fuel came from BWRs and 63% came from PWRs. However, 57% of the assemblies were from BWRs; 43% were from PWRs. Each assembly type has a unique combination of distinguishing characteristics, such as envelope dimensions, fuel rods per assembly, cladding type, and structural materials.

Initial enrichment and burnup can vary significantly with a single assembly type. For example, the first fuel loading of a reactor may have three sectors; one has an enrichment of 1.7%, the second an enrichment of 2.7%, and the third an enrichment of 3.7%. Discharge of the assemblies would be as follows:

- The first sector would be discharged at the end of the first cycle, with an average burnup of 10 GWDt/MTU;
- The second sector would be discharged at the end of the second cycle, with a burnup of 20 GWDt/MTU;
- The third sector would be discharged at the end of the third cycle, with a burnup of perhaps 30 GWDt/MTU.

Forty-five BWR assembly types were discharged from 41 BWR's between 1968 and 1992. However, five types made up 58% of the BWR assemblies discharged during this period. General Electric fabricated about 91% of the discharged BWR assemblies. As fabricated, most are 5.44 inches wide, 176.2 inches long, and contained 183 to 195 kilograms of uranium. Over this period (1968 and 1992), the average burnup was 22.3 GWDt/MTU. However, during the period of 1968 to 1972 the average burnup was only 5.9 GWDt/MTU; for the ten-year period 1968 to 1977, the average burnup increased to 10.1 GWDt/MTU. Some of this fuel had burnup as low as 0.3 GWDt/MTU.

Seventy-six PWRs discharged 47 PWR assembly types between 1968 and 1992. Five types make up 56% of the PWR assemblies. Westinghouse fabricated about 58% of the PWR assemblies discharged during this period. As built, most are 8.44 inches wide, 159.8 inches long, and contain 425 to 460 kg of uranium. Over this period, the average burnup was 30.9

GWDt/MTU. However, during the time period of 1968 to 1977, the average burnup was only 21.5 GWDt/MTU.

Typical physical characteristics of unirradiated LWR fuel assemblies are given in Table 1. Typical nuclear characteristics of LWR fuel assemblies are given in Table 2. The plutonium isotopic mix of various grades of plutonium is given in Table 3.

V. RADIATION LEVEL OF SPENT FUEL

Between discharge and one year, the gamma radiation falls by about a factor of 10, another factor of about 10 from 1 year to 10 years, and then falls roughly with the 30-year half-life of ^{137}Cs from 10 to about 300 years.

The Red Team[11] previously calculated the dose at 1.0 meter from the surface of both 10-year-old PWR and BWR fuel assemblies using the code MicroShield. The PWR assembly had an assumed burnup of 40 GWDt/MTU. At the axial midplane of the PWR assemble, the dose rate at the surface was calculated to be about 22,000 rem/hr and at 1 meter from the surface was about 1,400 rem/hr. At forty years of age, this dose rate would have dropped to about 700 rem/hr at 1 meter. However, average burnup of the spent fuel discharged between 1968 and 1992 was only 30.9 GWDt/MTU, about 75% of the value assumed by the Red Team. Although other isotopes do contribute to the dose, the ^{137}Cs isotopes provide the primary radiation dose. As a first approximation, the dose will be about 75% of the value calculated by the Red Team or about 16,500 rem/hr at the surface and about 1,050 rem/hr at 1 meter from the surface at tem years of age. At forty years, this dose rate would have dropped to about 525 rem/hr at 1 meter.

I Table 1. Typical Physical Characteristics of Unirradiated LWR Fuel Assemblies

	BWR^{ref}	PWR^{ref}
Overall assemble length, m	4.470	4.059
Cross section, cm	13.9 x 13.9	21.4 x 21.4
Fuel elements length, m	4.064	3.851
Active fuel height, m	3.759	3.658
Fuel element OD, cm	1.252	0.950
Fuel element array	8 x 8	17 x 17
Fuel rods per assembly	63	264
Assembly total weight, kg	275.7	657.9
Uranium/assembly, kg	183.3	461.4
UO2/assembly, kg	208.0	523.4
Zircaloy/assembly, kg	56.9	108.4
Hardware/assembly, kg	9.77	26.1
Total structural metal/assembly, kg	67.7	134.5

Table 2. Typical Nuclear Characteristics of LWR Fuel

Parameter	BWR	PWR
Uranium per assembly, kg		
Initial	183.3	461.4
Discharge	176.3	440.7
Enrichment, wt% ^{235}U		
Initial	2.75	3.20
Discharge	0.69	0.84
Pu per assembly at discharge, kg	1.57	4.32
Average power, MW/assembly	4.75	17.3
Average specific power, kw/kg initial uranium	25.9	37.5
Average discharge burnup, MWd/MT initial uranium	27,500	33,000
Irradiation duration, full-power days	1062	880

Table 3: Approximate isotopic composition of various grades of plutonium

Grade	Isotope				
	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
Weapons-grade	0.00012 0.0005	0.938 0.936	0.058 0.06	0.0035 0.004	0.00022 0.0005
Fuel Grade	0.001	0.861	0.12	0.016	0.002
Reactor-grade	0.013 0.01	0.603 0.62	0.243 0.22	0.091 0.12	0.050 0.03
MOX-grade (Com)	0.019	0.404	0.321	0.178	0.078
MOX-excess WG	0.02	0.54	0.227	0.151	0.062

In the Red Team calculation, the BWR assembly was assumed to have a burnup of 35 GWDt/MTU. At the axial midplane of the PWR assemble, the surface dose rate was calculated to be about 17,000 rem/hr and about 640 rem/hr at 1 meter at ten year of age. At forty years, this dose rate would have dropped to about 320 rem/hr at 1 meter. However, average burnup of spent fuel discharged between 1968 and 1992 was only 22.3 GWDt/MTU. This is only about 65% of the value assumed by the Red Team, so, the dose will be about 65% of the value calculated by the Red Team or about 11,000 rem/hr at the surface and about 420 rem/hr at 1 meter. At forty years this dose rate would have dropped to about 210 rem/hr at 1 meter.

During the period[10] 1968 to 1977, the average burnup was 10.1 GWDt/MTU. This is only 29% of the burnup assumed by the Red Team. The dose rate at the surface is about 5,700 rem/hr and 210 rem/hr at 1 meter. At forty years, this dose rate would have dropped to about 105 rem/hr at 1 meter.

During the time period[10] 1968 to 1972 some of the fuel had burnup as low as 0.3 GWDt/MTU and will be about 40 years out of the reactor at the midterm of the Immobilization Program. The dose rate from this aged spent fuel is estimated to be about 10 rem/hr.

V. CAN-IN-CANISTER IMMOBILIZATION OPTION

The Department of Energy (DOE) elected to pursue a duel path strategy for plutonium disposition[3]. One is irradiation as a mixed oxide (MOX) fuel in commercial light-water reactors. The second is immobilization in a ceramic material surrounded by high-level waste (HLW) glass encased in a stainless steel canister. A flowsheet for the process is given in Figure 1.

DOE has declared as excess to national security needs 38.2 tonnes of weapons-grade plutonium, and about 14.3 tonnes of combined fuel- and reactor-grade plutonium. DOE anticipates declaring an additional 7 tonnes to be excess[3]. Of this 59.5 about 7.5 tonnes of fuel-grade plutonium (primarily from N-Reactor) will be dispositioned as spent fuel at the Geologic Repository and it was estimated that ~ 2 tonnes will be declared below the safeguards termination limit and be discarded as TRU waste at WIPP. Part (< 18 tonnes) or all of the remaining 50 tonnes of excess plutonium will be immobilized in a ceramic waste form following the dual path disposition strategy.

The plutonium isotopic compositions of feed[3] stocks vary from 3% ^{240}Pu to about 40% ^{240}Pu. The assay varies from <10 wt% to >99 wt%. The last purification varies from the early 1960s to the late 1990s; therefore the ^{241}Am content is about 15-wt %. Actinide impurities include depleted uranium (DU), enriched uranium (EU), neptunium, thorium, and americium. The feedstock contains about 17 tonnes of DU; the amount of EU is uncertain but is expected to be between 0.6 and 2.6 tonnes. The Pu-to-U ratio varies from trace DU in the plutonium to trace plutonium in fully enriched U (93% ^{235}U, EU). The tramp impurities are dominated by the elements Al, C, Ca, Cl, Cr, Fe, F, Ga, K, Mg, Mo, Na, Si, Ta, W, and Zn.

To obtain a reasonably consistent plutonium feed[3], blending will be necessary (see figure 2). Blending large (50 kg Pu) batches, in two stages, will minimize processing and characterization costs, and will improve both product quality and reproducibility. Since EU will be introduced to the ceramic formulation

calculated as if it were plutonium, the amount of DU added will dilute the EU to below 20% in all cases. (This assures that the uranium could not be used for weapons without enrichment.) Each canister is expected to contain about 28 kg of plutonium. However, up to one-half of the plutonium could be replaced by EU with the final ^{235}U isotopic $\leq$20%. Tramp neptunium and thorium amounts in the plutonium feed have not been determined. Both will probably be only a few kgs to a few tens of kg. Therefore, in each canister, the actinide content may vary as follows:

Pu:	10 to 28 kg.
EU:	0 to 14 kg
DU:	$\geq$56 kg
Am:	200 ppm to 1.5 kg
Np:	0 to 2 kg
Th:	0 to 4 kg

Tramp impurities will also vary within loose limits - less than 50-wt % of the calculated plutonium content. Therefore, tramp impurities will be 14 kg per canister. The ceramic formulation contains hafnium and gadolinium neutron absorbers in atom ratios of about 1:1:1 = Pu:Hf:Gd.

VI. THE IMMOBILIZATION PROCESS

The plutonium Immobilization process can be divided into head-end, first-stage immobilization, and second-stage immobilization (See figure 1). These are discussed below.

The head-end will have the capability to:

- Convert metals to oxides
- De-jacket unirradiated fuel
- Grind materials
- Calcine materials
- Leach soluble salts from materials.

Large-scale blending will be used to minimize other processing and characterization costs and to improve product quality and reproducibility (see figure 2). This blending will smooth out the plutonium and uranium isotopics and therefore smooth out the heat load. It will also smooth out the tramp impurities to an acceptable level. As a result, the first stage immobilization process will have a reasonably consistent product.

The first stage of immobilization will convert plutonium oxide with its accompanying uranium oxide and impurity oxides into ceramic disks containing about 50-grams of plutonium and 100-grams of uranium will be welded inside stainless steel cans.

In the second stage of immobilization, cans of plutonium ceramic disks will be loaded inside empty canisters, 10 ft x 2 ft, and the canister will be filled with HLW glass at the Defense Waste Processing Facility (DWPF). The radiation from the HLW glass will provide a radiation deterrent similar to 30 year old spent fuel

The present thinking on how to encapsulate the cans of ceramic immobilized plutonium is shown in Figure 3. The cans are loaded into a cage of bars that run parallel to the surface of the canister. It would be very easy to join these cans together, probably by tack welding. This "magazine" is loaded into the canister and held in place by plates that are perpendicular to the bars. A mechanism locks the magazines into place. By designing the cans and the rods the proper thickness, any breach of the canister would cut the cans and shatter the ceramic. The shape and thickness of the plates would preclude cutting the plates during breach of the canister

Such designs require that the terrorist have to come into the radiation field to place burn bars on the rods and plates so that they could prevent the ceramics from shattering and the pieces from scattering. This would add both dose exposure and time to the attempted attack.

VII. RADIATION DOSE OF CAN-IN-CANISTER

The ^{137}Cs in HLW glass produced in DWPF will provide the radiation shield[4]. The amount of ^{137}Cs required was calculated using the MCNP code, a 3D Monte Carlo radiation transport code. For DWPF canisters, containing immobilized plutonium ceramics, the amount of glass will be about 1325 kg. (A glass-only DWPF canister will contain about 1680 kg of glass.) A dose rate of 100 Rem/hr one meter off the mid-plane of the

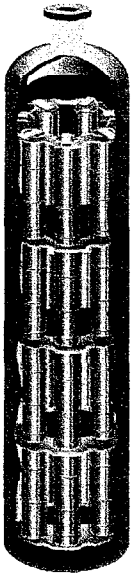

Figure 3. The can-in-canister design.

canister requires 5.36 kCi of ^{137}Cs. To assure a dose rate of 100 Rem/hr at one meter 30 years after fabrication, the curie loading at fabrication would be 10.8 kCi of ^{137} Cs.

The Savannah River Site[4] has made a number of waste work-off projections over the years. The 1996 projection averaged 13 kCi ^{137}Cs/canister. As there is some flexibility in the way the ^{137}Cs is processed, it would be fairly easy to assure at least 12 kCi ^{137}Cs per canister containing Pu. In fact, the process to prepare ^{137}Cs feed for the DWPF has been delayed; DWPF is currently producing "sludge only" glass containing only traces of ^{137}Cs. Depending upon the time required to start up the ^{137}Cs process, it may be possible to assure > 18 kCi ^{137}Cs/canister. Therefore, it may be possible to increase the dose rate, at fabrication, from 200 Rem/hr at one meter, to 400 Rem/hr.

VIII. ANALYSIS AND CONCLUSIONS

The external gamma radiation from the Pu-bearing canisters is comparable to that from 30 year old spent fuel assemblies, giving a comparable barrier to theft and diversion.

An analysis of the techniques and effort required by a clandestine group to remove the plutonium from the radiation field of 30 -year old spent fuel assemblies indicates that a clandestine process could be done without lethal doses of radiation to the workers.

Removing the cans of plutonium from the radiation field of the can-in-canister system without over-irradiating the people would be very difficult. Water shielding to lower the radiation dose, as is easily done for spent fuel assemblies, is not practical. Shadow shielding of the canister during the removal of the HLW glass from the canister would reduce the radiation dose received. However, sever radiation sickness and highly probable deaths within the clandestine group would be anticipated in about half of the scenarios studied. One could, of course, build robots to do the necessary work, but this is much greater sophistication than a simply cutting wheel that could be used to chop-up spent-fuel assemblies in a pool of water.

The shape and the weight of the HLW canister make the mechanical manipulation necessary for the canister far more difficult than the mechanical manipulations required for the spent fuel assemble.

Once the cans of plutonium ceramic are removed from the major radiation field, they would have to be decontaminated by some scheme. Again, this is time and dose to the clandestine group. This step is not necessary for spent fuel as the desired material is leached directly from the chopped fuel pieces. The disk would then have to be removed form the stainless steel cans and sized according to the requirement of the dissolution procedure developed by the clandestine group.

An analysis was made of flowsheets for the recovery of plutonium both from spent fuel assemblies and from canisters of immobilized plutonium. This analysis also indicated that it is more difficult to recover the plutonium from the HLW glass canisters than from spent fuel assemblies. Since the immobilized product contains a 2 to 1 mole ratio of uranium to plutonium and a 2 to 1 mole ratio of neutron absorbers to plutonium, full scale chemical processing and purification is necessary. The details of the chemical process for the recovery of plutonium from spent fuel have been published in numerous reports and books. Although flowsheets have been developed on a laboratory scale (only to demonstrate proof-of-principal) for the recovery of plutonium from the ceramic immobilized product, these proof-of-principal flowsheets have not been published in the unclassified literature. A terrorist group

might be able to develop the required procedures to recover the plutonium from the ceramic disks, but it would take both time and resources. The amount of time required would depend upon the plutonium processing expertise of the group developing the procedures and flowsheets.

A canister of plutonium-ceramic typically will contain 20 to 28 kg of plutonium, whereas a typical LWR fuel rod will contain about 4 to 5 kg of plutonium. From a proliferation standpoint, this would seem to favor the spent fuel, but spent fuel assemblies may be stored bundled together in casks, so that a comparison of a canister of immobilized plutonium to a cask of spent fuel assemblies may be more valid.

The proposed MOX fuel assemblies prepared from weapons-grade plutonium will contain about 28 kg of plutonium when fresh. After irradiation, they will contain about 20 kg of plutonium. Basically, the same holds true for MOX fuel assemblies for commercial reactors.

Theft of a single assembly or a few assemblies would require that the terrorist group provide their own shielding cask for transport of the assemblies to the processing site. Theft of a canister of immobilized plutonium would certainly require that the group provide a shielding cask for transport of the canister.

The canisters of plutonium will weigh about 2 tonnes, whereas typical fuel assemblies weigh one-third to three-quarters of a tonne. The greater weight of the canister favors the canister for theft resistance. Also, several LWR spent fuel elements would have to be stolen to obtain an equivalent amount of plutonium, or even to obtain sufficient plutonium to make a weapon.

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Figure 1. Pu Immobilization Three Stage Production Plant

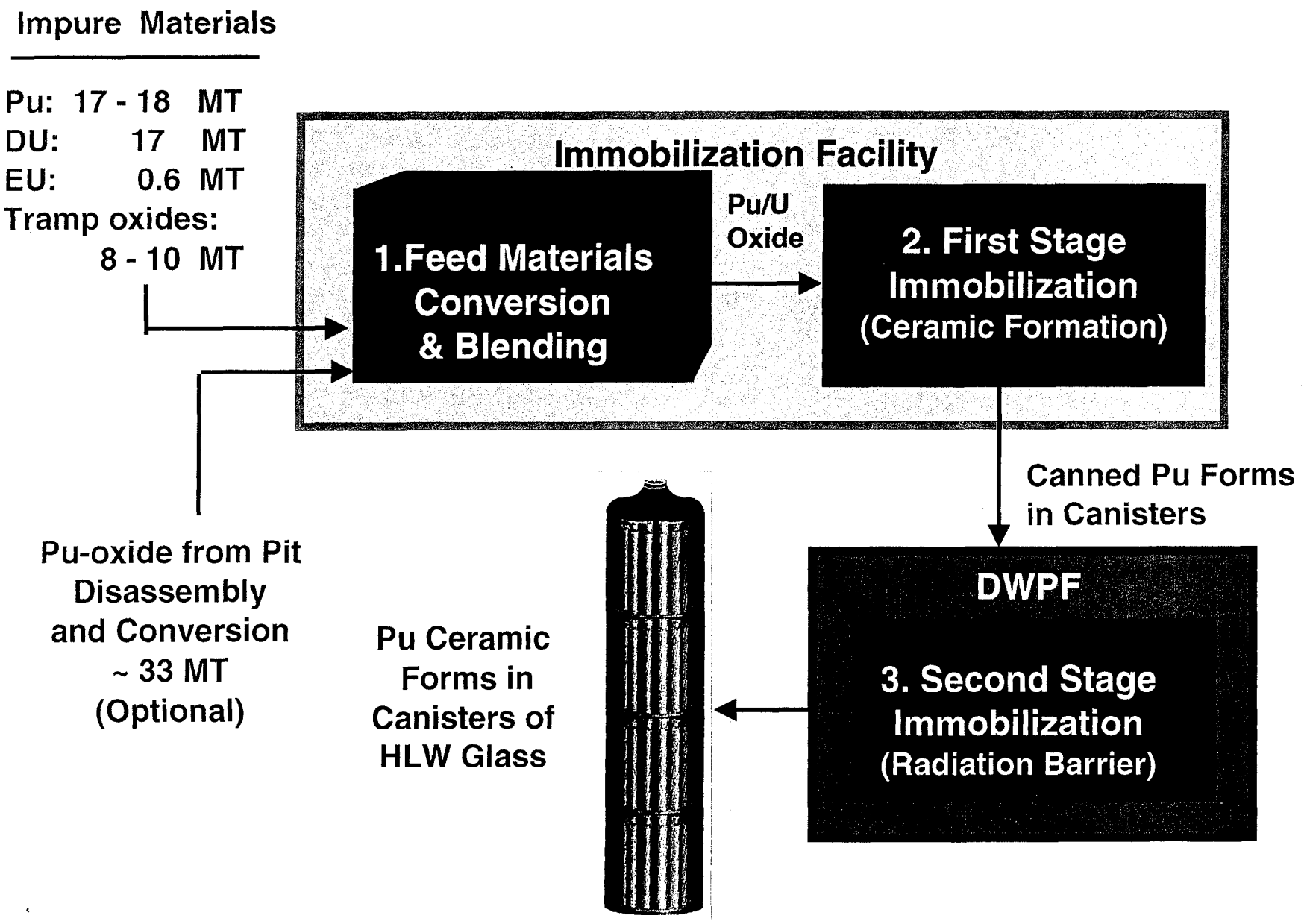

Figure 2. A two stage blending process concept